𝔄𝔯𝔤𝔬𝔫𝔫𝔢 𝔑𝔞𝔱𝔦𝔬𝔫𝔞𝔩 𝔏𝔞𝔟𝔬𝔯𝔞𝔱𝔬𝔯𝔶

ATTEMPTED PREPARATION OF
SINGLE CRYSTALS OF ALPHA PLUTONIUM -
A PROGRESS REPORT

by

Alan F. Berndt

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60440

ATTEMPTED PREPARATION OF
SINGLE CRYSTALS OF ALPHA PLUTONIUM -
A PROGRESS REPORT

by

Alan F. Berndt

A Report of Metallurgy Program 2.5.3

May 1964

TABLE OF CONTENTS

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

ATTEMPTED PREPARATION OF
SINGLE CRYSTALS OF ALPHA PLUTONIUM -
A PROGRESS REPORT

by

Alan F. Berndt

ABSTRACT

The standard methods of crystal growth are discussed in light of their applicability to the problem of growing single crystals of alpha plutonium. The results of some experiments designed to prepare single crystals of alpha plutonium are presented. These experiments include attempts at crystal growing by (1) solid-state reactions, (2) electrolytic reduction of plutonium halides, (3) precipitation from a liquid metal in convection cells, and (4) reduction of plutonium halides by lithium in nonaqueous solvents.

The indexing of powder patterns and the determination of the unit cell of alpha plutonium[1,2] were proved by an X-ray study of a small single crystal of a plutonium-neptunium alloy having the alpha-plutonium structure.

I. INTRODUCTION

It is extremely desirable, and often mandatory, to use single crystals in connection with studies of physical properties. Single crystals are not required for all types of measurements on materials with isotropic crystal structures since many properties are independent of crystallographic direction. Single crystals are important, however, in studies dealing with elastic and plastic properties and, also, where the effects of grain boundaries are significant. Most of the properties of materials with anisotropic crystal structures are functions of crystallographic direction, and require single crystals for their measurement.

The crystal structure of alpha plutonium is anisotropic (monoclinic).[1,2] In view of the current effort to determine the properties of this material, it would be advantageous to have single crystals of alpha plutonium available. A program has been initiated to achieve this goal. A method for preparing single crystals of alpha plutonium is not immediately

obvious for two reasons. (1) The standard solid-state techniques for grow-ing crystals of a metal, such as recrystallization and grain growth, strain anneal, etc., are unattractive for the growth of alpha plutonium because of the low temperature of the $\alpha \rightleftharpoons \beta$ transition, about 112°C. (2) Plutonium goes through five solid-state transformations upon cooling from the liquid to the temperature range of the alpha phase. The program is being pursued on the philosophy that single crystals might be grown by any method capable of yielding plutonium at a temperature within the stability range of the alpha phase.

This report discusses various methods of crystal growth, and the applicability of these methods to the problem of preparing single crystals of alpha plutonium, together with the results of several experiments. The various techniques of crystal growth can be divided into two major categories, based upon the source of the material being prepared in the form of a crystal. These categories are (1) homogeneous systems, wherein the source material has the same composition as the single crystal; and (2) heterogeneous sys-tems, wherein single crystals of a given composition are grown from a source material that contains other elements or compounds. Both categories include techniques involing a gaseous or liquid source material, and the first category also includes methods that involve source material in the solid state.

II. GROWTH FROM HOMOGENEOUS SYSTEMS

A. Gaseous Sources

Single crystals of some materials can be grown by direct condensa-tion of the vapor. A method based on this principle is unattractive for alpha plutonium. The data for the vapor pressure of plutonium between 1100° and 1500°C[3] extrapolate to 10^{-35} atm at 112°C. Extremely long periods of time would be required to produce an appreciable deposit. Since the lowest pres-sures conveniently obtainable in vacuum systems are of the order of 10^{-13} atm, it would be practically impossible to prevent contamination of the deposited plutonium by oxygen, nitrogen, water vapor, and/or other gases. Crystals grown at temperatures above 112°C would not have the structure of the alpha phase.

B. Liquid Sources

The most frequent method of preparing single crystals of metals de-pends upon the solidification of the liquid under controlled conditions. At one atm pressure, single crystals of alpha plutonium cannot be prepared in this manner because the epsilon phase is in equilibrium with the liquid at the melting temperature. Stephens has shown that the number of stable phases is decreased if the pressure is increased.[4] An extrapolation of his data

indicates that the alpha phase may be in equilibrium with the liquid at pressures above 50 kilobars. Therefore, it may be possible to grow single crystals of alpha plutonium from the liquid at high pressures.

C. Solid Sources

Single crystals can be grown within the solid state by several methods, involving the migration of a boundary through the specimen in a controlled manner. In the modified Bridgeman, or phase-transformation, method this boundary is the interface between the two phases. It may be possible to grow single crystals of alpha plutonium by properly controlling the conditions of cooling rate and temperature gradient for samples cooled from above 112°C. The success of this method and the quality of the single crystals that might be obtained are questionable considering similar work with uranium. Large single crystals of alpha uranium can be grown by this method, but they contain substructure which makes them undesirable for most investigations. Two factors which influence the success of this method are the volume change and the fraction of the absolute melting temperature (a rough measure of the mobility of the atoms) at the phase transformation. For uranium, these values are 1.1%[5] and 0.667,[6] respectively; for plutonium, these values are ~10%[1,2,7,8] and 0.43.[8] The values for plutonium, when compared with those for uranium, are in a direction that indicates that single cyrstals grown in this manner would be poorer in quality. Other important factors are the relative strengths of the two phases and the mechanism of the phase transformation. Beta plutonium deforms more readily than alpha plutonium,[9] which means that the strains that would result from the volume change at the phase transformation might be relieved by the beta phase, thereby minimizing the lineage structure in the alpha phase. The mechanism of the beta-to-alpha phase transformation is not yet well-understood. A diffusion-controlled mechanism is more advantageous than a shear mechanism when attempting to grow single crystals by the phase transformation. Zachariasen and Ellinger report[2] that it has not been possible to find a simple structural relationship between alpha and beta plutonium. This indicates that a major rearrangement of the atoms is involved in the $\alpha \rightleftharpoons \beta$ transition.

Other techniques of preparing single crystals from solid sources involve the migration of grain boundaries. The more important methods are: (1) strain-anneal, (2) grain growth, (3) secondary recrystallization, and (4) various combinations of these. Since little is known about the deformation and recrystallization behaviors of alpha plutonium, it is difficult to foresee, at this time, whether any of these methods will be successful.

Several attempts have been made to grow large grains of the alpha phase by slowly cooling samples from above the temperature of the $\alpha \rightleftharpoons \beta$ transformation. All of the samples have shown gross cracking, and none of these tests has been encouraging. Figure 1 shows the microstructure

of a specimen cooled in a carburized tantalum crucible in vacuo from 650°C (in the liquid phase) by lowering through a furnace at the rate of 2.5 cm/day (total cooling time from 650°C to room temperature: about 6 days). Specimens have also been cooled by reducing the furnace temperature at the rate of about 1°/hr with similar results. Figure 2 illustrates the smaller grain size of a specimen quenched from 375°C (in the delta phase) into Octoil-S. X-ray, back-reflection, pinhole patterns (patterns made with monochromatic radiation in a Laue camera) confirm the larger grain size in the slowly cooled specimen. The grain size of the slowly cooled specimens, however, is not much larger than that of as-cast material (not illustrated), and the quantity of subgraining has not been decreased significantly. The effects of the 10% volume change during the $\alpha \rightleftharpoons \beta$ transformation appear to make this approach impractical.
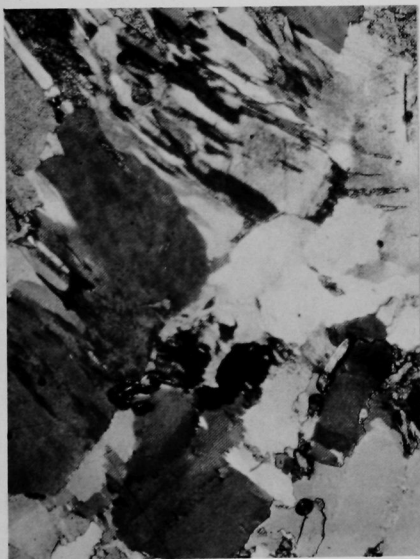
35780   250X   36311   250X

Figure 1. Plutonium Specimen Slowly Cooled by Lowering through a Furnace 2.5 cm/day. Cooling Time from 650°C to Room Temperature was about 6 Days. Polarized Light Illumination.
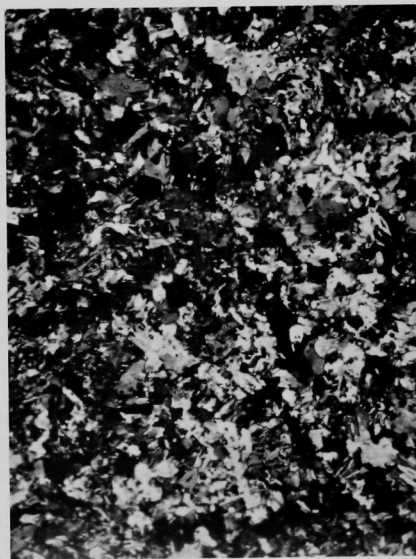
Figure 2. Plutonium Specimen Quenched from 375°C into Octoil-S. Polarized Light Illumination.

III. GROWTH FROM HETEROGENEOUS SYSTEMS

A. Gaseous Sources

Various metals can be obtained by thermal decomposition of volatile compounds. The compound is usually decomposed by a localized heat source (e.g., a hot wire which can also act as a seed), and the metal atoms are deposited on the hot surface. Under proper conditions of heat input and flow rate, single crystals can be grown. In the case of alpha plutonium, the high temperatures required to decompose plutonium compounds would seem to be incompatible with the stability of the alpha phase.

B. Liquid Sources

Single crystals are prepared from heterogeneous liquid systems by many methods. The three basic methods are (1) electrolytic reduction of ions, (2) deposition from a saturated solution by slow removal of the solvent, or by taking advantage of changes in solubility with temperature, and (3) precipitation as a result of a chemical reaction.

1. Electrodeposition. Plutonium metal can be produced by electrolysis of a plutonium salt in a suitable solvent. If the product is to be alpha plutonium, the solvent must melt below 112°C. Fused-salt solvents are unattractive because those that melt below 112°C must contain either potentially explosive nitrates or low-melting halides, such as aluminum trichloride, which are thermodynamically unstable with respect to plutonium metal.[10] Aqueous solvents appear unfavorable because plutonium reacts readily with water.

Electrolytic deposition experiments were performed in various organic solvents using several plutonium salts as electrolytes. The cathode was platinum, and the anode consisted of a piece of freshly cleaned (by filing) plutonium wrapped in a platinum wire. A six-volt storage battery was used as a power supply. Where conductivity was observed, the current was adjusted with a rheostat to a maximum of 0.1 ma to permit slow deposition. The results of some of the experiments are summarized in Table I.

Table I

SUMMARY OF ELECTROLYSIS EXPERIMENTS

Solvent	Solute	Is Salt Soluble?	Does Solution Conduct?	Was Cathode Deposit Formed?
Ethanol	$PuCl_3$	Yes	Yes	Yes
Ethanol	PuF_4	Slightly	Yes	No
Ethanol	$PuCs_2Cl_6$	Slightly	Yes	No*
n-Butanol	$PuCl_3$	Yes	Yes	Yes
Mineral Oil	$PuCl_3$	No	No	No
Pyridine	$PuCl_3$	Yes	Yes	No

* A saturated solution of $PuCs_2Cl_6$ in ethanol gave no cathode deposit, but formed a precipitate of PuO_2 upon electrolysis.

From these experiments the following generalization is drawn. The more polar the solvent, the greater the solubility of the plutonium salts and the greater the conductivity, and, unfortunately, the greater the tendency of the plutonium to react with the solvent.

Deposits were placed in thin-walled quartz capillaries and examined by the Debye-Scherrer technique. The deposits obtained from the electrolyses of $PuCl_3$ in ethanol and n-butanol consisted mainly of PuO_2; however, there were several spots on the Debye-Scherrer patterns at positions where lines from alpha plutonium should be. This suggested the existence of large grains of alpha plutonium in the capillary. One such capillary was broken; the largest piece in the deposit was isolated and mounted. The Debye-Scherrer pattern of this particle consisted only of about two dozen spots at positions corresponding to alpha-plutonium lines. This particle (about 0.1 x 0.1 x 0.01 mm) could possibly have been a single crystal of alpha plutonium. Attempts to prove this conjecture by orienting this crystal on a Weissenberg camera were unsuccessful when the spots became indistinct, probably because of the destruction of the crystal by oxidation.

2.   Convection Cells. Methods based upon the solubility of a material in a solvent are normally used to prepare single crystals of compounds. Ewald and Tufte[11] have used the temperature dependence of the solubility of tin in mercury to prepare single crystals of alpha tin (gray tin), which transforms to beta tin (white tin) at 13.2°C.[12] Their method uses a convection cell through which mercury flows under the influence of a thermal gradient. Tin is dissolved at the hotter portion of the cell and precipitates at the colder portion. Crystals about 1 cm³ in size have been grown in a month using a difference in solubility of the order of 0.08 a/o.

The success of this method depends upon the availability of a solvent for the phase whose crystal is to be grown. For alpha plutonium, alloys of liquid alkali metals appear to be the most suitable solvents, because they are the only known liquid metals in which alpha plutonium can be the solid phase in equilibrium with the liquid, although the solubilities may be extremely small.[13,14,15] The magnitude of the solubility is not as important as the differences in solubility with temperature. The primary difficulty would appear to be associated with contamination by oxygen. This can be kept to a minimum by controlling the purity of the alkali metals, and by adding lithium, which forms a more stable oxide than does plutonium.[10]

To investigate the possibility of applying this method to alpha plutonium, convection cells have been constructed. These cells are of two types. One is essentially identical to that described by Ewald and Tufte,[11] and the other is the "harp" design shown in Figure 3. In all systems investigated to date, the solvent has been a eutectic alloy of sodium and potassium (NaK) alloyed with about 1 a/o lithium and small additions of other

elements, in an attempt to increase the solubility of the plutonium without destroying the favorable phase equilibrium. Spectrographic analysis of the NaK is presented in Table II. The purity of the alloying elements is presented in Table III. The metallic plutonium was from Argonne batch CM-8, with analysis as given in Table IV.
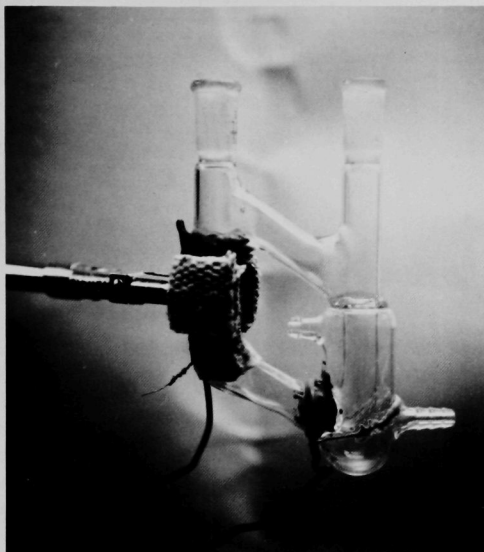
Figure 3
Convection Cell

38420

Table II

SPECTROGRAPHIC ANALYSIS
OF NaK

Element	Concentration
Al, Rb, Si	0.01% - 0.001%
Li, Mg	0.001% - 0.0001%
As, Hg, P, Sr, Zn	<0.1%
B, Ba, Bi, Ca, Co, Cr, Fe, Mn, Mo, Ni, Pb, Sb, Sn, Ti, V, Zr	<0.01%
Ag, Be, Cu	<0.001%

Table III

ANALYSIS OF
ALLOYING ELEMENTS

Element	Purity, by Weight
Hg	99.9999+%
In	99.9996%
Tl	99.977%*

*Impurities, 200 ppm Pb,
30 ppm Cu.

Table IV

ANALYSIS OF PLUTONIUM BATCH CM-8
(Expressed as Number of Impurity
Atoms per Million Plutonium Atoms)

Element	Concentration	Element	Concentration	Element	Concentration
C	141	Am	9*	Al	< 45
H	408	Cr	5	Co	< 4
N	242	Fe	30	K	< 6
O	151	Ga	3	Mo	< 5
Si	60	Li	0.3	Na	< 21
		Mn	1	Pb	< 1
		Ni	4		
		U	29		
		W	7		

* Extrapolated to date of electrolysis.

The convection-cell experiments are summarized in Table V.
In all cases (except cell 4) the plutonium charge, after being cleaned by
filing, was broken under NaK to provide fresh surfaces to facilitate solution.
These surfaces were wetted, and they remained clean under unalloyed NaK
for extended periods of time. As a check on the solubility, small amounts
of two solutions were withdrawn with a medicine dropper, passed through a
sintered glass filter at room temperature, and analyzed for plutonium. The
results were as follows:

0.44 μg/cc in NaK

0.50 μg/cc in NaK 1 w/o Hg.

Unfortunately, it is not known whether these values are true indications of
solubility, as opposed to entrapped solid particles, nor is the temperature
known.

Table V

SUMMARY OF CONVECTION CELL EXPERIMENTS

Cell	Type	Charge	Solvent[a]	Temp (°C) Hot	Cold	Time (days)	Appearance of Charge at End
1	Ewald	Pu	NaK	68	58	26	Shiny
2	Ewald	Pu	NaK 1 w/o Hg	114	77	60	Dull black
3	Harp	Pu	NaK 1/2 w/o Tl	150	90	102	Shiny
4	Harp	Pu-82 w/o Tl	NaK	116	73	49	Broken up
5	Harp	Pu	NaK 5 w/o In	(b)	98	63	Dull black
6	Harp	Pu	NaK 5 w/o In	159	94	195	(c)

a In all cases, small amounts of lithium have been added to getter oxygen.

b Thermocouple failed

c Currently in operation.

The charge from cell 4 was broken up but did not go into solution. The charges from cells 1, 2, and 5 showed no evidence of attack, other than the observed blackening of the charges from cells 2 and 5. The charge from cell 3 appeared to have been slightly rounded at the broken faces, suggesting some solubility. The solvent from cell 3, after decomposition by ethanol, produced a residue containing thallium and any plutonium that went into solution. This residue was examined by the Debye-Scherrer method. In addition to alpha thallium, there was a trace of a phase which, if plutonium-containing, could be identified only as PuB_6. The boron may have originated in the pyrex walls of the cell, or in the ethanol used to decompose the NaK.

Attempts at growing single crystals of alpha plutonium by using convection cells are being continued even though the above results are not encouraging. No concrete evidence of solution of the charge or precipitation of crystals has been obtained. This method is theoretically feasible whenever there is a temperature dependence of solubility, although the length of time required may be prohibitive because of the low solubilities encountered.

3. <u>Reduction of Plutonium Halides</u>. According to the values of the published free energies of formation,[10] the alkali metals are capable of reducing plutonium fluorides or chlorides to metallic plutonium. If an inert solvent for both a plutonium halide and an alkali metal can be found, then the reduction can be made to proceed in solution at temperatures below the $\alpha \rightleftharpoons \beta$ transition with the precipitation of plutonium metal. In a suitably designed container the reaction may proceed slowly, through diffusion, with the growth of crystals.

A suitable solvent may be either liquid ammonia or a solution of triphenyl boron (TPB) in diethyl ether. Unfortunately, ammonia, which is gaseous at room temperature, must be cooled or pressurized to liquify. TPB has the advantage that it is liquid at room temperature and atmospheric pressure.

Lithium dissolves in TPB to produce a black solution which reacts with plutonium salts. One such reaction product was a black powder which turned green on exposure to the box atmosphere. The Debye-Scherrer pattern of the green material was that of PuO_2, suggesting that the original reaction product may have been finely divided metallic plutonium.

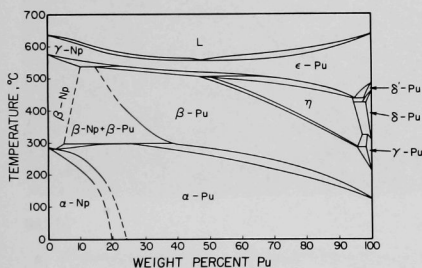Further experiments, diffusion-controlled, are in progress. The plutonium halides currently being investigated are $PuCl_3$, PuF_4, and $PuCs_2Cl_6$. Several reaction products from these tests have yielded complex Debye-Scherrer patterns which have not been completely analyzed. Two of these patterns showed spots at positions where lines from alpha plutonium should be, and were almost identical in appearance to patterns obtained from some electrolytic residues (see Section III-B-1, page 8).

IV. PLUTONIUM-NEPTUNIUM ALLOY STUDIES

The feasibility of growing single crystals of alpha plutonium by solid-state reactions can be enhanced if the number of solid phases between the liquid and the alpha phase can be reduced. This may be accomplished either by applying pressure,[4] or by alloying with neptunium.[16] In the latter case, the phase obtained will not be pure alpha plutonium, but it can have the alpha-plutonium structure. In view of the expected greater ease of preparing single crystals of this alloy, this line of investigation has been pursued. This alloy may be considered to be homogeneous in the sense that any crystals grown will have the same composition as the bulk melt.

The equilibrium phase diagram for the plutonium-neptunium system, according to Mardon et al.,[16] is shown in Figure 4. Alloys of plutonium and neptunium having a composition of more than 24 a/o Pu exist as the alpha-plutonium structure at room temperature (~25°C), with lattice constants only slightly smaller (~0.7% at 24 a/o Pu) than those observed for pure alpha plutonium.[16] For compositions between about 39 a/o Pu and 48 a/o Pu, the only solid phases that are reported to exist are the epsilon plutonium (gamma neptunium), beta plutonium, and alpha plutonium phases, and the temperature of the $\alpha \rightleftharpoons \beta$ transition is raised to about 270°C.

38630

Figure 4. Plutonium-Neptunium Constitution Diagram [From Mardon et al.[16]]

A melt of 43.4 w/o Pu-56.6 w/o Np was prepared in vacuo in a carburized tantalum crucible, cooled at 1/2°/hr from the liquid to 265°C, held for five weeks, cooled at 1°/hr to 70°C, and furnace cooled. Back-reflection pinhole patterns of this melt showed it to consist of extremely large grains, in fact the largest grains of any sample investigated. This is confirmed by the metallographic evidence presented in Figure 5.

Figure 6 is a back-reflection Laue pattern of this specimen taken with a 10-mil collimator centered near the position of the arrow on Figure 5a, near the center of an apparent grain. This is a single-crystal pattern, with lineage structure clearly evident.

The crystal structure of alpha plutonium metal has been determined entirely from X-ray powder-diffraction data.[1,2] Because of the low symmetry of this structure (space group, P_{2_1}/m) and the resulting complexity of the powder patterns, it would be of value to have single-crystal verification of the unit-cell dimensions.

36170   75X   36171   75X

a. 0° rotation of stage. The arrow indicates the   b. 45° rotation of stage.
   area examined by the X-ray, back-reflection,
   Laue technique.

Figure 5. Microstructures of Pu-56.6 w/o Np Alloy Electropolished in a Bath of 32 Parts Orthophosphoric Acid,
           Nine Parts Water, and 59 Parts Ethoxy-Ethanol. Polarized Light Illumination.

Figure 6

Back-reflection Laue Pattern
of Pu-56.6 w/o Np Alloy

38488

Since this ingot was brittle, a single-grain particle (about 0.02 x 0.02 x 0.01 mm) was fractured from it, isolated, and mounted for X-ray examination. This crystal was oriented by means of oscillation photographs to rotate about a 19.37 Å axis. Rotation and Weissenberg photographs showed that the unit cell was monoclinic (Laue symmetry, C_{2h}) with

$$a_0 = 6.12 \pm 0.02 \text{ Å}, \quad b_0 = 4.80 \pm 0.01 \text{ Å},$$
$$c_0^! = 19.37 \pm 0.026 \text{ Å}, \quad \beta' = 146.39 \pm 0.13°.$$

By a transformation of axes these can become

$$a_0 = 6.12 \pm 0.02 \text{ Å}, \quad b_0 = 4.80 \pm 0.01 \text{ Å},$$
$$c_0 = 10.95 \pm 0.04 \text{ Å}, \quad \beta = 101.74 \pm 0.23°.$$

Considering shrinkage due to alloying, these are the same (within experimental error) as the published unit-cell dimensions of alpha plutonium.[1,2] The observed intensities are in good qualitative agreement with those calculated from the published atomic positions.[17]

This result therefore provides single-crystal proof of the indexing of the powder data and determination of the unit cell of alpha plutonium, so ingeniously performed by Zachariasen and Ellinger.

To look for crystallographic evidence of a low-temperature phase transformation, this crystal was held for 67 hours at liquid-helium temperature. No change in the diffraction pattern was observed after warming to room temperature.

V. CONCLUSIONS

Various methods for single-crystal preparation as applied to the problem of growing single crystals of alpha plutonium have been discussed. The most promising methods seem to be those based upon growth from a heterogeneous liquid system at temperatures within the stability range of the alpha phase.

The most encouraging results have been from experiments in the plutonium-neptunium system, which show that large grains having the alpha-plutonium structure can be produced. A small particle fractured from one of these grains has been used to provide single-crystal proof of the indexing of the powder data and determination of the unit cell of alpha plutonium.

VI. ACKNOWLEDGMENTS

REFERENCES

1. Zachariasen, W. H., and Ellinger, F., Crystal Structure of Alpha-Plutonium Metal, J. Chem. Phys., 27, 811-812 (1957).

2. Zachariasen, W. H., and Ellinger, F., The Crystal Structure of Alpha-Plutonium Metal, Acta Cryst., 16, 777-783 (1963).

3. Wilkinson, W. D., Extractive and Physical Metallurgy of Plutonium and Its Alloys, Interscience Publishers, New York (1960), p. 4.

4. Stephens, D. R., The Phase Diagram of Plutonium, J. Phys. Chem. Solids, 24, 1197-1202 (1963).

5. Chiotti, P., Klepfer, H. H., and White, R. W., Lattice Parameters of Uranium from 25 to 1132°C, Trans. ASM, 51, 772-786 (1959).

6. Blumenthal, B., The Transformation Temperatures of High Purity Uranium, J. Nucl. Mat., 2, 23-30 (1960).

7. Zachariasen, W. H., and Ellinger, F. H., The Crystal Structure of Beta Plutonium Metal, Acta Cryst., 16, 369-375 (1963).

8. Zachariasen, W. H., Crystal-Structure Studies of Plutonium Metal, Chapter X, The Metal Plutonium, A. S. Coffinberry and W. N. Miner, eds., The University of Chicago Press, Chicago (1961), pp. 99-107.

9. Gardner, R. H., and Mann, I. B., Mechanical Properties and Formability Studies on Unalloyed Plutonium, Plutonium 1960, E. Grison, W. B. H. Lord, and R. D. Fowler, eds., Cleaver-Hume Press, Ltd., London (1961), pp. 513-570.

10. Glassner, A., The Thermochemical Properties of the Oxides, Fluorides, and Chlorides to 2500°K, ANL-5750, (1957).

11. Ewald, A. W., and Tufte, O. N., Gray Tin Single Crystal, J. Appl. Phys., 29, 1007-1009 (1958).

12. Van Lent, P. H., The Position of Gray Tin in the Tin-Mercury System, Acta Met., 9, 125-128 (1961).

13. Waldron, M. B., Phase Diagrams of Plutonium Alloys Studied at Harwell, Chapter XXI, The Metal Plutonium, A. S. Coffinberry and W. N. Miner, eds., The University of Chicago Press, Chicago (1961), pp. 225-239.

14. Schonfeld, F. W., Plutonium Phase Diagrams Studied at Los Alamos, Chapter XXII, The Metal Plutonium, A. S. Coffinberry and W. N. Miner, eds., The University of Chicago Press, Chicago (1961), pp. 240-254.

15. Schonfeld, F. W., Plutonium Phase Diagrams Published by the Russians, Chapter XXIII, The Metal Plutonium, A. S. Coffinberry and W. N. Miner, eds., The University of Chicago Press, Chicago (1961), pp. 255-264.

16. Mardon, P. G., Pearce, J. H., and Marples, J. A. C., Constitution Studies on the Neptunium-Plutonium Alloy System, J. Less-Common Metals, 3, 281-292 (1961).

17. Berndt, A. F., and Lloyd, L. T., Tabulations of Calculated Crystallographic Data for Alpha Plutonium, ANL-6676 (June 1963).

15. Jackson, R. C., Palmer, ...

16. Baroch, A. ...